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**METHOD AND DEVICE FOR CLEANING AUTOMOBILE INDOOR AIR**

[Jidosha Shitsunai Kuki no Joka Hoho Oyobi Sochi]

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AUTOMOBILE INDOOR AIR

(Completely revised specification of

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Specification

Clean copy of specification (no changes in contents)

1. Title of the invention

Method and device for cleaning automobile indoor air

2. Patent Claims

1. A method for cleaning automobile indoor air characterized by the fact that outdoor air and/or circulated indoor air is transmitted, by a suction gas exhaustion mechanism, through an acidic gas removal filter layer, an activated carbon filled layer, a heat exchanger, and a carbon monoxide removal filter layer in proper order.

2. A device for cleaning automobile indoor air constituted by an acidic gas removal filter, activated carbon, a heat exchanger, and a carbon monoxide removal filter obtained by adhering a noble metal catalyst to a honeycomb-shaped structure.

3. Detailed explanation of the invention

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<sup>1</sup> Numbers in the margin indicate pagination in the foreign text.

The present invention concerns a method for removing harmful gases from automobile indoor air and for maintaining the resulting cleaned state as well as a device for the same.

In recent years, hazards of atmospheric pollutions have become extremely conspicuous, and in particular, hazards caused by automotive exhaust gases are immense. The respective concentrations of carbon monoxide, hydrocarbons, nitrogen oxides, etc. within exhaust gases are /2 being regulated today, and although attempts are being made to improve designs of engines and/or to apply contact-type oxidation or reduction catalysts as concrete pollution preventive measures, fully satisfactory results have yet to be obtained. Even in a case where a previously developed technique for ensuring the regulated exhaust gas values is available, furthermore, it is difficult to avoid the arising of a local harmful gas atmosphere with extremely high pollutant concentrations during traffic jams in metropolitan areas or during travels through tunnels. Such a fact suggests that actual operators and passengers of automobiles are constantly exposed to dangers, which cannot be overlooked from the standpoint of health management. Of all the harmful gases, the hazards of carbon monoxide are especially conspicuous, for it leads not only to health hazards ascribed to the formation of a fair amount of CO hemoglobin within the blood stream of an operator, etc. in the midst of a massive traffic flow but also to the deteriorations of operating abilities, which may each in turn contribute to traffic accidents. Narrow automobile interiors are

normally ventilated based on the intake of outdoor air, but since the air outside the automobile has become polluted at present, not even a ventilating operation can be carried out in a satisfactory fashion. In a case where the automotive interior has become filled with polluted air upon the opening of a door and/or window, furthermore, harmful gases should be detoxified immediately.

In other words, the preventions of the emissions of these toxic gases from generation sources have been taken into consideration in the prior art, as evidenced by the regulations on exhaust gas concentrations, etc., but conversely, the health aspects of operators and co-passengers may be said to have been relatively overlooked. A precedent which may be said to reflect, though vaguely, the latter concept concerns a method wherein indoor air is circulated and distributed through an adsorption layer of activated carbon, etc. via a compact fan for deodorizing the air. At present, however, neither products nor proposals are known with regard to the removal and/or detoxification of carbon monoxide, which is most hazardous from the standpoint of health management.

The present inventors have endeavored, against the foregoing backdrop, not only to develop an oxidation catalyst which is capable of inhibiting the concentration of carbon monoxide within indoor air at or below a certain environmental standard value but also to build an apparatus for said catalyst, as a result of which the present invention has become completed.

In other words, one objective of the present invention is to provide a method which is capable of lowering the concentrations of harmful gases, above all carbon monoxide, included in either outdoor air taken into the interior for ventilating purposes or indoor air to low and safe levels. Another objective is to provide a device for cleaning automobile indoor air which is capable of     /3  
lowering the concentrations of harmful gases to low and safe levels.

These objectives can be achieved, as Figure 1 shows, by the transmission of outdoor air and/or circulated indoor air, by a suction gas exhaustion mechanism, through an acidic gas removal filter layer, an activated carbon filled layer, a heat exchanger, and a carbon monoxide removal filter layer in proper order.

The present inventors probed conditions necessary for efficiently detoxifying carbon monoxide within the aforementioned outdoor air or circulated indoor air, as a result of which it was discovered that it is necessary to satisfy at least the following three conditions. In other words, such conditions are (1): That a high-activity catalyst which can be oxidized at a relatively low temperature of 100°C or below be used; (2): That, prior to the arrival of the air scheduled to be cleaned at the catalyst filter, matters which serve as catalyst poisons at or below 100°C, namely gaseous sulfurous acid, nitrogen dioxide, halogenated compounds, dust and dirt, hydrocarbons (e.g., oil mist, etc.), and other organic compounds, etc., be removed; and (3): That the embodied

apparatus be compact enough to be applied to existing automobiles and be accompanied by minimal pressure losses. These three conditions will be explained.

First, as far as the essentiality of oxidizing and removing carbon monoxide at a low temperature of  $100^{\circ}\text{C}$  or below is concerned, a thermal energy arising auxiliarily from an existing automobile should be used from the standpoint of cleaning the air without imposing any burden whatsoever on the driving power of the automobile, and a method wherein hot water retrieved from an engine cooling radiator is sampled as a heat source may, for example, be said to be desirable in this context. It is therefore necessary to select a catalyst which exhibits a sufficiently high activity at  $100^{\circ}\text{C}$  or below even in a state characterized by a severe temperature variation. The present inventors [1 character inserted] investigated catalysts endowed with low-temperature activities in acknowledgment of this requisite, as a result of which catalysts of platinum group elements such as platinum, palladium, etc. were determined to be ideal.

Next, it was ascertained that these platinum group element catalysts become gradually deactivated by microscopic quantities of acidic gases included in the air, and as a result of investigations of potential countermeasures, adsorbents such as activated carbon-supported manganese oxide, ion exchange materials, etc. were discovered to be effective. At a temperature of  $100^{\circ}\text{C}$  or below, furthermore, the catalyst becomes significantly deactivated as a result of the adhesions not only of these acidic

gases but also of all matters normally included in the air,  
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namely dust and dirt, oil mist, and other organic compounds, and therefore, it is necessary to remove these substances. General filters are not sufficient for achieving this goal, but rather, activated carbon is effective, and one the surface area of which is at least  $500 \text{ m}^2/\text{g}$  and the average micropore diameter of which is  $10 \sim 200 \text{ \AA}$  is especially desirable to use.

Thirdly, automobiles are designed to maximize the occupant space, and accordingly, accessory devices are compactly configured within a narrow space, due to which only a severely limited space is likely to be available for installing a cleaning device anew, and thus, as compact a device as possible that can be applied to all vehicular types must be designed. There are, on the other hand, many variable elements (e.g., carbon monoxide [concentration], air temperature, operative state, etc.), and their variances are also significant. It is necessary for the rate of air permeated for cleaning purposes to correspond inevitably to such variations. In a case where a cooling function is being invoked, for example, the absolutely minimal volume of outdoor air needed for ventilation should be cleaned, whereas in a case where the indoor carbon monoxide concentration is high, it is necessary to lower the indoor carbon monoxide concentration to a safe level by quickly cleaning circulated indoor air or outdoor air. The optimal volume of air thereby treated for cleaning purposes varies significantly depending on prevailing



circumstances [1 character deleted], and a catalyst filter and a device accompanied by minimal pressure losses should be orchestrated for coping with such contingencies. It was discovered as a result of investigations of methods and devices which were hoped to satisfy the aforementioned three conditions that satisfactory results can be obtained only in a case where the method discussed in detail below is orchestrated.

First, noble metal catalysts such as platinum and palladium are suitable as the carbon monoxide oxidation catalyst, as has been mentioned earlier. In order to induce an unhindered progress of a carbon monoxide oxidation reaction over an extended period of time in the presence of a noble metal catalyst of the prior art, however, a temperature of 200°C or higher is normally necessary, and therefore, it cannot be used as it is for achieving the objectives of the present invention. The present inventors proceeded to study methods for preparing catalysts endowed with oxidation activities at relatively low temperatures, as a result of which a catalyst that met said objectives was obtained by impregnating or coating a carrier (e.g., aluminum silica, silica-alumina, etc.) [1 character deleted] with a solution of a noble metal compound such as chloroplatinic acid, palladium chloride, etc., by reducing the obtained structure at a temperature of 200°C or below, and by then thermally treating it in an oxygen-containing atmosphere (e.g., air, etc.) within a temperature range of 120 ~ 180°C. In a case where air is used, however, care must be taken in        /5

that the catalyst activity may decrease unless coexisting dirt and dust and other organic compounds, etc. are removed.

The reduction may, furthermore, be carried out based either on a dry method which uses hydrogen, etc. or on a wet method which uses formalin, etc., but in either event, it is essential for the temperature to be concomitantly designated at 200°C [or below]. It is indispensable, furthermore, for the thermal treatment temperature to be strictly regulated, whereas a duration of at least 1 hour, preferably at least 10 hours, virtually suffices as the treatment period. In a case where the thermal treatment temperature drops below the lower limit of the stipulated range, the durability becomes inferior, whereas in a case where it exceeds the upper limit of the same, the activity significantly decreases [1 character deleted], as a result of which [the obtained catalyst] becomes unusable in the method of the present invention.

The activity of the noble metal catalyst which has been prepared according to the foregoing procedures, however, gradually decreases in the presence of acidic gases (led by gaseous sulfurous acid), dirt and dust, oil mist, and other organic compounds within polluted air scheduled for cleaning, and therefore, it is necessary to remove such substances prior to their arrival at the catalyst filter. It has come to be known that the uses of activated carbon and acidic gas removal agents are effective as countermeasures in this context. The "acidic gas removal agent" collectively encompasses substances which are

capable of adsorbing and removing acidic gases based on chemical adsorption or physical adsorption, and it is instantiated by anion exchange resins, anion exchange fibers, activated carbon, activated carbon fiber, etc. Both high-basicity and low-basicity types are usable as the anion exchange resin, although it is more desirable to use the high-basicity type in consideration of the removal effect. It was also discovered to yield results more favorable than those ascribed to so-called "MR-type resins" with large surface areas, and corresponding methods were previously proposed.

As a result of subsequent research, however, it was discovered that these resins often exert undesirable effects on the noble metal catalyst. An ion exchange resin on which manganese has become adsorbed, in particular, is an extremely effective agent for removing gaseous sulfurous acid, but at the same time, it is undesirable in that it often releases potentially poisonous substances in relation to the noble metal catalyst.

In other words, the most desirable method is a method which uses a structure obtained /6 by supporting manganese oxide on activated carbon, and such a matter can be obtained by impregnating activated carbon with an aqueous potassium permanganate solution and by then drying the obtained adsorbed structure.

Ordinary activated carbon, furthermore, is effective for removing other matters, namely dirt and dust, oil mist, and various other organic compounds, and as such, a grade the particle

size of which is the smallest possible without entailing an excessive pressure loss and which is equally endowed with a high capacity for removing the oil mist, etc. may be adventitiously selected, although one with a specific surface area of  $500 \text{ m}^2/\text{g}$  or larger and an average micropore diameter of  $10 \sim 20 \text{ \AA}$  is ideal in each case. As far as the third condition, namely a compact catalyst filter with a minimal pressure loss, is concerned, the use of a granular catalyst of the prior art is utterly inappropriate, and instead, it is desirable to use a honeycomb-shaped structure to which a catalyst has become adhered [16 characters deleted, 12 characters deleted]. The "honeycomb-shaped structure" signifies an aggregate of micropores with cross-sectional areas of approximately  $1 \sim 25 \text{ m}^2$ , and so-called "honeycomb"-shaped structures with void ratios of approximately  $20 \sim 90\%$  suffice, whereas there are no special restrictions on its constituent material and micropore cross-sectional shape. In other words, as (a) ~ (d) in Figure 2 illustrate, ones with square, triangular, modified triangular, hexagonal, or various other shapes may be used without any inconveniences, although it is most advantageous to use hexagonal ones. It is desirable, however, for the cross-sectional areas of these micropores to be confined to a range of  $1 \sim 25 \text{ m}^2$  in the context of embodying a compact device, and a range of  $2 \sim 10 \text{ m}^2$  is especially desirable. Next, it is desirable for the void ratio to be designated within a range of  $20 \sim 90\%$ , although the material thickness becomes limited depending on the type of said material and on the manufacturing

method, and therefore, a range of 30 ~ 80% is sufficiently usable. The overall shape may, furthermore, be arbitrarily selected in consideration of installation methods, and rectangular parallelepiped, cubic, and cylindrical shapes, etc. are conceivable, as (e) and (f) in Figure 2 indicate.

Next, not only ceramics but also glasses and other inorganic compounds and organic compounds [1 character deleted] such as plastics are usable as constituent materials, which represents one peculiar characteristic of the present invention. The reason is because the reaction can be induced at 100°C or below.

Methods for adhering noble metal catalysts to these honeycomb-shaped structures are instantiated not only by a method wherein a carrier such as alumina, silica, silica-alumina,

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diatomaceous earth, etc. is adhered to a honeycomb, wherein the adhered structure is impregnated with a solution of a noble metal salt, and wherein the obtained impregnated structure is reduced but, in the case of a material with a relatively low heat resistance such as plastics, etc., also by a method wherein a catalyst is preliminarily adhered to the aforementioned carrier, which prevails as a powder or microscopic granule, and wherein the obtained adhered structure is further adhered to a honeycomb-shaped structure by using an adhesive, etc., and either method can be effectively orchestrated.

A material with a high heat resistance (e.g., cordurite, etc.) may, for example, be immersed in an aqueous dispersion of

alumina hydrate, and after the adhered structure has been dried and then baked while the temperature is being gradually elevated to a maximum of 600°C, preferably 500°C; it is immersed in and impregnated with a noble metal solution and then reduced. An acid treated honeycomb-shaped structure of a glass wherein the internalization ratio of the silica component is at least 90%, furthermore, exhibits a high heat resistance, and it can be used as a catalyst filter according to similar procedures.

Next, in a case where a catalyst has become adhered to a honeycomb-shaped structure with a relatively low heat resistance (e.g., plastics, etc.), it is advantageous to adhere, by using an adhesive, a carrier (e.g., alumina, etc.) to which a catalyst has already become adhered. Optimal adhesives differ depending on the constituent material of the honeycomb and the constituent material of the carrier, although the following are generally usable: Epoxy adhesives, polyvinyl acetate adhesives, polyacrylic acid ester adhesives, rubber adhesives, polystyrene adhesives, polyvinyl chloride adhesives, polyvinylidene chloride adhesives, polyvinyl ether adhesives, polyvinyl acetal adhesives, phenol resin adhesives, urethane resin adhesives, unsaturated polyester adhesives, polyvinyl alcohol adhesives, polyacrylamide adhesives, and cellulose adhesives. After such an adhesive has been adhered to the honeycomb-shaped structure, the adhesion of the catalyst is induced by an appropriate method [e.g., one wherein the aforementioned carrier-supported catalyst is scattered by the sand blast format, one wherein the same is homogeneously adhered

electrostatically by impressing a high voltage (30,000 ~ 150,000 V), one wherein the honeycomb-shaped structure to which the adhesive has become adhered is immersed in an inert gas stream characterized by a fluid state, etc.].

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Figure 3 shows a magnified view of the catalyst-adhered honeycomb-shaped structure obtained according to these procedures, and in this embodiment, a state where the catalyst-supporting carrier is adhered to the micropore interior of the honeycomb is shown.

Such a method can also be applied to an acidic gas filter. In other words, in a case where an activated carbon on which manganese oxide is being supported is pulverized into a size of 5, preferably 10, mesh or less and where the obtained product is adhered to a honeycomb-shaped structure according to the aforementioned procedures, a filter [1 character deleted] the pressure loss of which is far lesser than that of a case where granular carbon or pulverized carbon is directly used can be obtained.

Next, the aforementioned requisites will be explained further with reference to figures. Figure 1 shows an example of the cleaning device. Outdoor air or circulated indoor air is introduced into Cleaning Box No. 1 (2), via the dust removal filter (1), by the suction gas exhaustion mechanism (8), and after acidic gases, led by gaseous sulfurous acid, have been removed from it by the acidic gas removal filter layer (3), other residual

matters such as dirt and dust, oil mist, etc. are removed from it by the activated carbon filled layer (4), and the obtained air is then introduced into Cleaning Box No. 2 (5). The air thus introduced is heated by the heat exchanger (6), which uses hot water as a medium, at 50 ~ 100°C, and after it has been induced to arrive at the carbon monoxide removal filter (7), carbon monoxide is fortified [sic: Presumably "removed"], and the cleaned air is then introduced indoors. Ones of honeycomb-shaped structures are the most desirable as the acidic gas removal filter layer (3) and the carbon monoxide removal filter (7), which possesses the catalyst, in consideration of the necessity of minimizing the pressure loss. The air treatment rate presumably coincides with the minimal threshold necessary for normal ventilation, namely approximately 0.1 m<sup>3</sup>/min. to 5 m<sup>3</sup>/min., although it, too, represents a variable factor depending on the types of vehicles, and an appropriate value can adventitiously be selected by configuring switch and/or adjustment mechanisms. Ones made of various constituent materials such as papers, cloths, non-woven fabrics, etc. can be used as the dust removal filter (1). Cleaning Box No. 1, furthermore, serves a function of removing harmful components based mainly on adsorption, and it is desirable for it to be configured on a site characterized by the lowest possible temperature.

As for the heat source for the heat exchanger (6), the circulation of cooling water for a radiator represents the simplest method, although all other auxiliary heat sources of



automobiles can be used effectively. In other words, it is also possible to use [the heats of] the engine mainframe, exhaust gas pipe, etc.

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Next, as far as methods for actually configuring the present device on an automobile are concerned, it may, for example, be housed within an engine room, and a pipe configuration that enables the use of the radiator cooling water as a heat source is orchestrated, as Figure 4 shows. It is also possible to configure, in adjustable forms, a valve which enables the cleaning of either outdoor air taken in for ventilation purposes and circulated indoor air. As Figure 5 indicates, furthermore, it is extremely rational to release cleaned air indoors via the heater (9) or cooler (10) that the automobile possesses, and such a method is desirable.

In a case where the cleaning device and method of the present invention discussed above are actually applied to an automobile, not only carbon monoxide included in the air taken into the vehicular interior during its operation but also acidic gases (e.g., gaseous sulfurous acid, nitrogen dioxide, etc.), oil mist, and other organic compounds can be removed, based on which adverse effects exerted on humans by them can be avoided. Moreover, pleasant driving experiences are ensured since odors, dirt and dust, etc. are also removed. In the following, explanations will be provided with reference to application examples.

### Application Example 1

After commercially sold activated carbon "Shirasagi G" (manufactured by Takeda Yakuhin Kogyo Co.; 4 ~ 6 mesh) had been immersed in an aqueous solution which contained 0.4 mole/L of potassium permanganate at a bath ratio of 1 : 10 at room temperature over a 24-hour period, it was washed with water and then dried at 100°C, as a result of which an activated carbon-supported manganese oxide in which approximately 7  $\mu$ mole/g (activated carbon) of metallic manganese was retained was obtained. After approximately 1 kg of said activated carbon had been filled into a cleaning box, an air stream which included approximately 50 ppm of gaseous sulfurous acid was permeated through it at a rate of 1 m<sup>3</sup>/min. Although the contact period with said filter was only approximately 0.02 sec., nearly 100% of the gaseous sulfurous acid had become removed. Incidentally, the gaseous sulfurous acid concentration was measured by using Model GR-2C (atmospheric SO<sub>2</sub> measurement device manufactured by Denki Kagaku Keiki Co.).

### Application Example 2

After a cordurite honeycomb with a micropore (square shape) cross-sectional area of approximately 3 m<sup>2</sup> and a wall thickness of approximately 0.3 mm (overall shape: Cylinder with a diameter of approximately 93 mm and a height of 75 mm; void ratio: Approximately 72%) had been immersed in an aqueous dispersion of an alumina hydrate gel, it was thermally treated at 120°C for 5

hours, at 250°C for 4 hours, and at 500°C for 3 hours, as a result of which approximately 15% /10 of alumina became adhered with respect to the honeycomb weight. Next, the obtained structure was immersed in a 0.5 N aqueous hydrochloric acid solution which included 2.5% of palladium chloride and thus impregnated with palladium chloride, and after it had subsequently been reduced by using an aqueous solution which contained [8]% of formalin (pH: 10), it was thermally treated at 150°C for approximately 20 hours while a gaseous mixture of 20% of oxygen and 80% of nitrogen was being permeated.

An air stream which included approximately 50 ppm of carbon monoxide was heated at [80] ~ [85]°C and then transmitted through said honeycomb at a flow rate of 0.3 m<sup>3</sup>/min., as a result of which the carbon monoxide concentration dropped to 1 ppm or below.

### Application Example 3

After [300] g of the activated carbon-supported manganese oxide obtained in Application Example 1 and 500 g of activated carbon "Pittsburgh CAL" (manufactured by Organo Co.; 10 ~ [3]2 mesh) had been filled into Cleaning Box No. 1 shown in Figure 1, a cylindrical filter was attached to its distal end, and the obtained structure was installed in the lower frontal portion of an automobile. The radiator cooling water of its engine room, furthermore, was orchestrated to become partially circulated through a compact heat exchanger, whereas Cleaning Box No. 2 was embodied by configuring the honeycomb used in Application Example

2 immediately behind [said heat exchanger], and it, too, was installed in the engine room. Cleaning Box Nos. 1 and 2 were mutually linked via a flexible pipe, and the air emitted from Cleaning Box No. 2 was induced to flow into the automobile interior. A test was conducted by configuring the gas suction device shown in Figure 4 between Cleaning Box Nos. 1 and 2 in such a way that the outdoor air would flow inward at a rate of approximately 0.[8] m<sup>3</sup>/min.

In a case where the automobile was driven without activating the heat exchanger of Cleaning Box No. 2, the concentration of carbon monoxide included in the outdoor air that had flowed into the automobile interior was 5 ~ 100 ppm, whereas the concentration was lowered to 1 ppm or below as a result of the activation of said heat exchanger. The concomitant air temperature of Cleaning Box No. 2 was 80 ~ 90°C.

This outdoor air, furthermore, included approximately 6 ppm of gaseous sulfurous acid and 2 ppm of nitrogen dioxide, whereas the air which had been transmitted through Cleaning Box No. 1 included no more than 0.1 pphm of gaseous sulfurous acid and no more than 1.2 pphm of nitrogen dioxide.

Next, the passenger car in which said device had been installed was driven over an extended period of time for testing its durability, as a result of which absolutely no variations of the cleaning performances were observed even after a driving distance of 2,000 [km]. In a case where the car was driven after

the filter and Cleaning Box No. 1 had been removed, however, 5 ~ 20 ppm of /11 carbon monoxide came to migrate after a driving distance of merely 300 km.

#### Application Example 4

The activated carbon used in Application Example 1 was initially pulverized, and after a 150 ~ 200 mesh component had been selected, it was immersed in an aqueous potassium permanganate solution, and an activated carbon-supported manganese oxide was obtained according to similar procedures. Palladium, on the other hand, was adhered to a 50 ~ 200 mesh activated alumina according to procedures similar to those in Application Example 2.

A plastic honeycomb with a square (side dimension: 2.5 mm) micropore cross-sectional shape (wall thickness: Approximately 0.3 mm; overall shape: 75 x 75 x 30 mm rectangular parallelepiped) was coated with a thin layer of an epoxy adhesive, and 9 units of the aforementioned honeycombs to which activated carbon and alumina had been adhered were prepared. A constitution identical to that of Application Example 3 except that the granular activated carbon-supported manganese oxide of Application Example 3 had been substituted with said honeycomb-shaped filter and that the cordurite honeycomb had been substituted with 9 units of said catalyst-adhered plastic honeycomb was installed within a passenger car. An indoor circulation system, however, was orchestrated as an air flow path, and after the interior of the

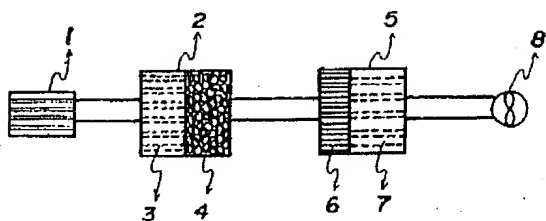
passenger car (volume: Approximately 23 m<sup>3</sup>) had been filled with air which included 100 ppm of carbon monoxide, the cleaning device was activated at a treatment rate of 0.3 m<sup>3</sup>/min., as a result of which the concentration of the carbon monoxide dropped to 1 ppm or below 10 min. later. Incidentally, the concentration of the carbon monoxide was measured by using APMA-10 (atmospheric carbon monoxide measurement device manufactured by Horiba Mfg. Co.).

#### 4. Brief explanation of the figures

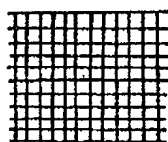
Figure 1 shows one constitutional example of the device of the present invention, whereas Figure 2 shows a morphological example of the honeycomb-shaped structure, whereas Figure 3 shows a state where an activated carbon-supported manganese oxide or carrier-supported noble metal metal has become adhered to such honeycombs. Figures 4 and 5 show configurational examples of cases where the devices of the present invention are installed in automobiles.

Applicant: Kanebo Co., Ltd.

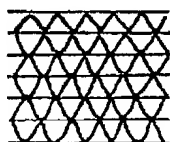
Agent: Koichi Minaguchi, patent attorney



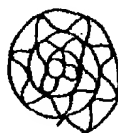
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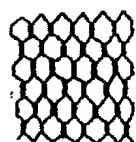
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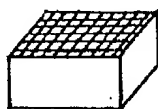
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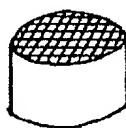
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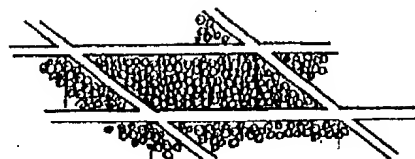


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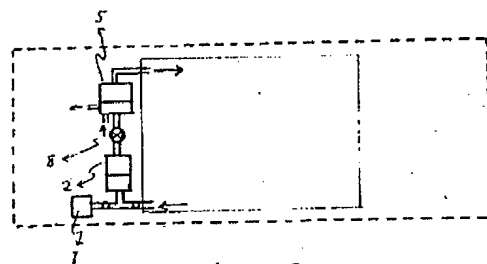


(f)

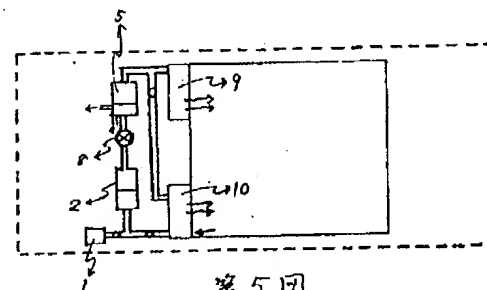
第 2 圖



第 3 圖



第 4 圖



第 5 圖

## 6. List of attached documents

- |                             |        |
|-----------------------------|--------|
| (1): Specification:         | 1 copy |
| (2): Drawings:              | 1 copy |
| (3): Application duplicate: | 1 copy |
| (4): Letter of attorney:    | 1 copy |

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END



Procedural Amendment Report (spontaneously submitted)

October 7, Sho 48[1973]

Dear Mr. Hideo Saito, Chief of the Patent Agency:

1. Display of the case

Japanese Patent Application No. Tokugan Sho 48[1973]-87115

2. Title of the invention

Method and device for cleaning automobile indoor air

3. Amending party

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5. Number of inventions added as a result of amendments: None

6. Contents of amendments

"Clean copy of specification (no changes in contents)"



**公開特許公報**

特許國 ( ) (6)



特許庁長官 斎藤英雄 殿

1. 発明の名称  
ジドウ シヤシツナイクウキ ジョウカホウホウ ソウチ  
自動車室内空気の浄化方法及び装置
2. 特許請求の範囲に記載された発明の数 2
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明細書(内容に変更なし)

1. 發明の名称

置装び及花方化浄の歌謡区柳博家血

## 2. 特許請求の範囲

(1) 外氣及び、又は循環室内空気を吸排気機構により、酸性ガス除去フィルター層、活性炭充填層、熱交換器、及び一酸化炭素除去フィルター層に順次通気せしめる事を特徴とする自動車室内空気の浄化方法。

④酸性ガス除去フィルタ—活性炭、熱交換器、及びヘニカム状構造物に貴金屬触媒を付着せしめた—酸化炭素除去フィルタ—から構成して成る自動車室内空氣の淨化装置。

## 8. 説明の詳細を説明

本発明は、自動車室内空気を有害ガスを除去し、清浄化された状態に維持するための方法及びその装置に関する。

近時大気汚染による被害が激増しており、特に、自動車排気ガスによる被害はますますひどい。今日排気ガス中の一酸化炭素、炭火

アや窓の開放により室内が汚染空気で充満された場合は、速やかに有害ガスを無害化すべきである。

即ち、従来より排気ガスの濃度規制等に見られる如く、発生源からこれら有害ガスを防止する事は考慮されてきたが、逆に運転者や乗客の健康は見過されてきた感がある。敢てこの思想に近い事例を取り上げるならば、室内空気を小型ファンを介して活性炭等の吸着層に循環流通せしめ脱臭を行なう方法がある。しかしながら、最も健康管理上実施しなけれなければならぬ一酸化炭素の除去無害化に關しては、製品はおろか提案もなされていない現状である。

本発明者等はこの点に鑑み室内空気の一酸化炭素濃度を環境基準値以下に抑制すべく酸化触媒の開発及び該触媒の装置化を探究し、本発明を完成したものである。

即ち、本発明の目的は換気として室内へ取入れる外気及び又は、室内空気中に含有される有害ガス、とりわけ一酸化炭素を安全な低濃度域まで減少せしめる方法を提供す

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水素、窒素酸化物等の濃度規制がなされ、その防止対策として具体的にはエンジンの改良、接触酸化又は、還元触媒の適用等が試みられているものの、未だ不完全な現状である。又、例え排気ガス規制値に達する技術が開発された場合に於いてさえ、自動車台数の増加に伴い、都市部に於ける交通渋滞時やトンネル内の走行時等には、局部的には、非常な高濃度の有害ガス雰囲気になる事は避け難い。この事は、実際に自動車運転する者や同乗者は常に危険にさらされている事を意味し、健康管理上見逃すわけにはいかない。とりわけ有害ガスの中でも一酸化炭素による被害は大きく、大量の交通流の中での運転者は血液中にかなりのCOヘモグロビンが形成されることによる健康障害はもとより、運転能力の低下を引起し、ひいてはそれが交通事故につながる恐れもある。通常、狭い車内の換気は外気を取り入れることによりなされているが、車外の空気が汚染されている現状では換気さえも満足に行えない有様である。又、ド

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るにある。他の目的は、有害ガスを安全な低濃度域に迅速減少せしめる自動車用空気浄化装置を提供することにある。

該目的は、第1図に示す如く、外気及び、又は循環室内空気を吸排気機構により、酸性ガス除去フイルター層、活性炭充填層、熱交換器、及び一酸化炭素除去フイルター層を順次通気せしめる事により達成される。

本発明者等は上述した外気及び又は室内循環空気中の一酸化炭素を効率よく無害化するための必要条件を追求した結果、少なくとも次の8条件を満たす必要があることを見出した。即ちその条件とは(1) 100°C以下の比較的低温で酸化可能な高活性触媒を使用すること。(2) 浄化するべき空気が触媒フイルターに達する前に、100°C以下に於いて触媒層となる亜硫酸ガス、二酸化窒素、ハロゲン化合物等及びその他の塩埃、オイルミスト等の炭化水素類、その他の有機化合物等を除去し去ること。(3) 既存の自動車に適用できるコンパクトな装置であり、且つ圧力損失が小さい装置であることとの以

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上8点である。

先ず100°C以下の低温で一酸化炭素を酸化除去する必然性は、自動車が行くため動力に何ら負担をかけずに浄化する必然性から、既存の自動車に付属する熱エネルギーを用いるべきであり、例えばエンジン冷却用のラジエーターから熱水を取り出し、それを熱源とする方法が望ましいと言えらる。従つて、100°C以下で且つ温度変化が激しい状態に於いても充分高い活性を有する触媒が必要である。本発明者はこの点に着目し、低温活性を有する触媒を検討した結果、白金触媒、例えば白金、パラジウム等が適していることを知つた。

次にこれら白金触媒は、空気中に微量に含まれる酸性ガスにより徐々に活性を失ひ去るを知り、その防止対策を検討した結果、活性炭担持マンガン酸化物や、イオン交換体等の吸着剤が有効であることを見出した。更に100°C以下の温度では、これら酸性ガスのみならず、通常の空気中に含まれる全ゆる塩埃、オイルミスト、その他の有機化

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合物の付着によつても著しく活性を失うため、これら物質を除去しておく必要がある。この目的に対しては一般のフィルターでは充分ではなく活性炭が有効であり、特に表面積が500 $\text{cm}^2/\text{g}$ 以上であり、平均細孔径が10~200 $\text{\AA}$ の物が好適に用いられる。

第8に自動車は居住空間を如何に広くするか工夫が要らざれており付属装置は狭い空間にコンバクトに配置されているので、浄化装置を新たに設置できる場所は極く限られており、何れの車種にも適用出来るには極力コンパクトな装置とすべきである。又、一酸化炭素、空気温度、走行状態等変動要素が多く、且つ変動幅も大きい。その変動に対し、浄化すべき風量も必然的に対応させる必要がある。例えば、クーラーを使用している場合に於いては、換気に必要な最低限度の量の外気を浄化すべきで、又、室内の一酸化炭素濃度が高い場合には、室内循環空気か、或いは外気を迅速に浄化し、室内の一酸化炭素濃度を安全な濃度迄減少させる必要がある。かように浄化すべき如

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理風量は状況に応じ、著るしく変動するものであり、それに対応するためには、圧力損失が小さい触媒フィルター及び装置とすべきである。上記8条件を満たす方法及び装置を検討した結果、次に詳述する方法を採用することにより、始めて満足な結果を得ることが出来た。

先ず一酸化炭素酸化触媒としては、前述の通り白金、パラジウムの如き貴金属触媒が適している。然しながら、従来の貴金属触媒では、一酸化炭素酸化反応を円滑に且つ長期間に亘つて進行せしめるには、通常200°C以上の温度が必要であり、このままでは本発明の目的に対して使用することは出来ない。しかるに本発明者等は、100°C以下の比較的低温度に於ける酸化活性を有する触媒の調整法に關し研究を重ねた結果、貴金属化合物、例えば塩化白金酸、塩化パラジウム等を溶解し、アルミナ、シリカ、シリカアルミナ等の担体に含浸又は被覆せしめた後、200°C以下の温度で還元し、次いで酸素含有雰囲気、例えば空気中にて

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到達する前に斯かる物質を除去する必要がある。この防止対策として活性炭及び酸性ガス除去剤を用いる事が有効であることを知った。酸性ガス除去剤とは、酸性ガスを化学的吸着、或いは物理的吸着により吸着除去し得る能力を有する物の総称であり、例示するならば、陰イオン交換樹脂、陰イオン交換繊維、活性炭、活性炭繊維等がある。陰イオン交換樹脂に於いては、強塩基型或いは弱塩基型の何れも適用可能であるが、除去効果を考慮するならば強塩基型が好ましい。又、表面積の大きい所謂MR型樹脂に比して好結果を与えることを見出し、これら方法については既に提案した。

しかるにその後行なつた研究の結果、これらの樹脂は、貴金属触媒に対して好ましくない影響を与える場合のあることを見出した。特にマンガンを吸着せしめたイオン交換樹脂は、亜硫酸ガスの除去には極めて優れた薬材であるが、同時に貴金属触媒に対し、被毒性の物質を放出することがあり、好ましくない。

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120~180°Cで熱処理を施すことにより目的に合致する触媒を得る事ができた。但し、空気を利用する場合に存在する塵埃その他の有機化合物等を除去しておかないと触媒活性がむしろ低下する事さえ起る場合があるので注意が必要である。

又、還元は水素等による乾式法、及びホルマリン等による湿式法の何れも好適に用いる事が出来るが、その場合の温度が200°Cで行なり事が肝要である。又、熱処理温度も厳密に調節する事が必須であり、処理時間については実質1時間以上、好ましくは10時間以上行なえば充分である。熱処理温度が該範囲の下限をはずれた場合は耐久性に乏しく、又、上限を越えた場合は活性が著しく低下するため本発明方法に用いる事は出来なくなる。

斯して調整された貴金属触媒を以つてしても浄化すべき汚染空气中に亜硫酸ガスを始めとする酸性ガス及び塵埃、オイルミストその他の有機化合物が存在するとその活性は除々に低下するため、触媒フィルターに

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れるものではない。即ち第2図の(向)～(四)に示されている様に、正方形、三角形、菱形、三方形、六角形、或いはその他の形状の何れであつても何ら障害をえないが、六角形のものが最も有利である。但しこれら細孔の断面積はコンバクトな装置とするためには1～25 $\mu^2$ の範囲内にあることが望ましく特に2～10 $\mu^2$ であるものが好適に用いられる。次に空間率は20～90%のものが望ましいが、材質及び製造方法により材質の厚みが限定されるため、通常80～80%のものであれば充分に利用し得る。又、全体の形状としては第2図の(向)、(四)の如く、直方体、立方体、或いは円柱形等、設置方法によつて任意に選べば良い。次に材質としてはセラミックスのみならず、ガラス或いはその他の無機化合物、更にはプラスチックの如き有機化合物でも用いる事ができる事は本発明の特徵の1つである。即ち100°C以下で反応できるためである。

これらハニカム状構造物に貴金属触媒を付

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即ち、最も望ましい方法としてはマンガン酸化物を活性炭に担持せしめたものを用いる方法であり、この物は過マンガン酸カリウム水溶液に活性炭を浸漬し、吸着せしめた後乾燥して得ることができる。

又、他の塵埃、オイルミスト等種々の有機化合物の除去に対しては、通常の活性炭が有効であり、圧力損失が過大にならない限り粒度の小さい物で、オイルミスト等の除去能力の高い物を適宜選ぶ事ができるが、何れの場合も比表面積が500 $\mu^2/g$ 以上、平均細孔径が10～20Åの物が適している。

第8の要件である圧力損失の小さいコンバクトな触媒フィルターは、従来の粒状触媒の使用は全く不適当であり、ハニカム状構造物に、触媒を付着せしめたものが適している。~~ハニカム状構造物に、触媒を付着せしめたものが適している。~~ハニカム状構造物とは、断面積1～25 $\mu^2$ 程度の細孔の集合体であり、空間率20～90%程度である所謂ハニカム状の構造を有するものである。材質及び細孔の断面形状により限定さ

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る場合は接着剤を用いて触媒の既に付着しているアルミナ等の担体を接着するのがある。接着剤の選定は、ヘニカムの素材、及び担体の素材等によつて異なるが一般には、エポキシ系接着剤、ポリ酢酸ビニル系接着剤、ポリアクリル酸エステル系接着剤、ゴム系接着剤、ポリスチレン系接着剤、ポリ塩化ビニル系接着剤、ポリ塩化ビニリデン系接着剤、ポリビニルエーテル系接着剤、ポリビニルアセタール系接着剤、フエノール樹脂系接着剤、ウレタン系接着剤、不飽和ポリエステル系接着剤、ポリビニルアルコール系接着剤、ポリアクリル酸アミド系接着剤、セルロース系接着剤等が使用可能である。ヘニカム状構造物にこれら接着剤を付着せしめた後、前記担体付触媒を、サンドブラスト法で振りかけるか、或いは高電圧(8万~15万ボルト)をかけ、静電気により均一に付着せしめるか、或いは不活性ガスにより流動化状態にし、この中に接着剤を付着せしめたヘニカム状構造物を浸漬せしめる等の方法によつて触

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着せしめる方法としては、アルミナ、シリカ、シリカーアアルミナ、珪藻土その他の担体をヘニカムに付着せしめた後に貴金属塩の溶液に浸漬、含浸せしめた後に還元する方法、或いはプラステックス等の比較的耐熱性の低い材質の場合には粉末乃至微小粒状の前記担体に先ず触媒を付着せしめた時、ヘニカム状構造物に接着剤等を用いて付着せしめる方法等があり、何れも効果的に採用する事が出来る。

例えば、コージュライト製ヘニカムの如き耐熱性の高い材質の場合には、アルミナ水和物の水分散液に浸漬、付着せしめた後、乾燥し、徐々に昇温しながら高くととも600℃、好ましくは500℃で焼成した後貴金属塩溶液に浸漬、含浸後還元する。又、ガラス製のヘニカム状構造物を酸処理し、シリカ成分を90%以上にしたものの耐熱性が高く、同様の方法によつて触媒フィニターとして利用することが出来る。

次にプラステックス等の比較的耐熱性の低いヘニカム状構造物に触媒を付着せしめ

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気は熱水を媒体とする熱交換器6によつて50~100°Cに加熱された後、一酸化炭素除去フイルター7に達し、一酸化炭素を強化し、清浄化された空気を室内へ導入する。酸性ガス除去フイルター8、及び触媒を有する一酸化炭素除去フイルター7は圧力損失を極力抑える必要上、ハニカム状構造物が最も優れている。処理風量は通常換気に必要な最低限度の0.1 m<sup>3</sup>/分乃至5 m<sup>3</sup>/分程度と考えられるが、これも車種による変動要因であり、切換、調節機構を設けて適宜選択出来る様考慮出来る。除塵フイルター1は紙製、布製、或は不織布等で種々の素材の物を利用することが出来る。又、浄化箱(1)は主として吸着によつて有害成分を除去するものであり、出来るだけ温度の低い箇所に設置することが望ましい。熱交換器6の熱源としてはラジエーターの冷却水を循環せしめる方法が最も簡便であるが、その他の自動車に付随する全ての熱源を有効に利用することができる。即ちエンジン本体、或は排気ガスパイプ等をも利

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媒を付着せしめることが出来る。斯くして得られた触媒付ハニカム状構造物を拡大して示したものが第8図であり、ハニカム細孔内に触媒担持体が付着している様子を示している。

又、該方法は酸性ガスフイルターについても応用することが出来る。即ちマンガン酸化物を担持した活性炭を5、好ましくは10メッシュ以下とした後、上述せる方法でハニカム状構造体に付着せしめて用いるれば、粒状炭又は破砕炭そのままで用いるよりも遙かに圧力損失の小さいフイルターを得ることが可能となる。

次に上述の要件を図面を以つて更に説明する。第1図に浄化装置の一例を示す。外気及び又は室内循環空気は吸排気機構8によつて除塵フイルター1を通して浄化箱(1)2に導入され、酸性ガス除去フイルター層8によつて亜硫酸ガスを始めとする酸性ガスを除去し、更に残留するその他の塵埃、オイルミスト等を活性炭充填層4で除去した後浄化箱(1)5へ導入する。導入された空

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# 実 施 例 1

市販の活性炭「白鷺G」(武田薬品工業製 4~6メッシュ)を浴比1:10の割合で過マンガン酸カリウム0.4mol/lの水溶液に室温下で24時間浸漬した後、水洗100℃で乾燥して金属マンガンを換算して約7mmol/g(活性炭)を保持する活性炭担持マンガン酸化物を得た。該活性炭約1kgを浄化槽に充填し、亜硫酸ガス約50pphmを含有せしめた空気を1m<sup>3</sup>/minの風量で流した。該フイALTERとの接触時間は約0.02秒であつたが、亜硫酸ガスはほぼ100%除去されていた。なお亜硫酸ガス濃度は電気化学計器製大気中SO<sub>2</sub>測定装置GR-20形を用いて測定した。

# 実 施 例 2

細孔断面積が約8m<sup>2</sup>の正方形で、壁の厚さ約0.8mm、全体の大きさは直径約93mm、高さ75mmの円柱形で空間率約72%のコージェニライト製ハニカムをアルミナ水和物グル

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用可能である。

次に本装置を実際に自動車に配置する方法としては、例えば第4図の如くエンジンルーム内に格納し、ラジエーターの冷却水を熱源として利用する様配管する。又、換気用に取り入れられる外気及び室内循環空気の何れをも浄化出来る様バルブを設け調整可能な形に出来る。或は、第5図の如く、車体に装備されたヒーター9又はクーラー10を通して清浄空気を室内へ放出する様にすれば極めて合理的であり、好ましい方法である。

上述した如き本発明による浄化装置及び方法を自動車に適用し、実施する事により、走行中に車内に入る空気に含まれる一酸化炭素はもとより、亜硫酸ガス、二酸化窒素等の酸性ガス、更にはオイルミスト、他の有機化合物等が除去され、これらによる人体への悪影響を阻止する事が可能となる。又、悪臭、その他塵埃も除去され、快適なドライブを可能ならしめるものである。以下実施例によつて説明する。

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℃で4時間、500℃で8時間熱処理し、ヘニカム重量に対し約15%のアルミナを付着せしめた。次いで塩化パラジウム2.5%塩酸0.5規定水溶液に浸漬し、塩化パラジウムを含浸せしめた後PH10のホルマリン8%水溶液で還元し、150℃で約20時間酸洗20%、塩素80%のガスを渡しながら熱処理した。

一酸化炭素約50ppmを含有せしめた空気を80~85℃に加熱し、0.8ℓ/minの流量に調整して該ヘニカムを通過せしめた所、一酸化炭素は1ppm以下となつた。

### 実施例 3

実施例1で得た活性炭担持マンガン酸化物300gと、活性炭「ビツツバーグCAL」(オムガノ製10~82メッシュ)500gを第1図の浄化箱(1)に充填し、その先に円筒形のフィルターを装着し、自動車の前方下部に設置した。又、エンジンルームのラジエーター冷却水の一部を小型熱交換器を循環する様にし、そのすぐ後方に実施例2で用

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いたヘニカムを装着して浄化箱(2)を作成し、エンジンルームに設置した。浄化箱(1)と(2)をフレキシブルパイプで連結し、浄化箱(2)を出た空気を室内に流入する様にした。外気が流入する量が約0.8ℓ/minとなる機器4図の如く吸気装置を浄化箱(1)と(2)の間に設置してテストした。

浄化箱(2)の熱交換器を作動させずに走行させると、室内に流入してくる外気に含まれている一酸化炭素濃度は5~100ppmであつたが、熱交換器を作動させると1ppm以下となつた。この時の浄化箱(2)の空気温度は80~90℃であつた。

又、この外気には約6ppmの亜硫酸ガス及び2ppmの二酸化窒素が含まれていたが、浄化箱(1)を通過した後には亜硫酸ガスは0.1ppm以下、二酸化窒素が1.2ppm以下になつていた。

次に該装置を装着した乗用車を長期に亘つて走行し、耐久性をテストした所、2000km走行後でも浄化性能には何ら変化はなかつた。しかし、フィルター、浄化箱(1)を外

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例3と同様の構成で乗用車に設置した。但し空気の流通経路は室内循環系とし、乗用車の室内(容積約28 $\text{m}^3$ )を一酸化炭素100ppmを含む空気で満たした後、0.8 $\text{m}^3/\text{min}$ の処理量で浄化装置を作動させた所、10分後には一酸化炭素の濃度は1ppm以下に減少した。なお一酸化炭素の濃度は漏れ製作所製大気中一酸化炭素測定装置APMA-10を用いて測定した。

#### 4.図面の簡単な説明

第1図は本発明装置の1構成例を示し、第2図はハニカム状構造物の形状例を示し、第3図はこれらハニカムに活性炭担持マンガン酸化物或いは担体付貴金属触媒が付着している様子を示したものである。第4図及び第5図は本発明装置を自動車に装着する場合の配置例を示す。

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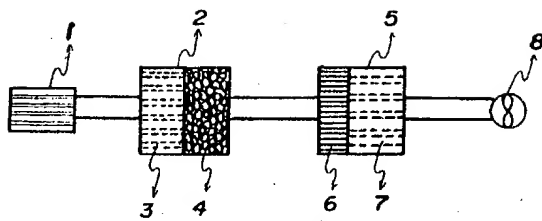
して走行した所、僅か300 $\text{m}$ の走行によつて5~20ppmの一酸化炭素が流入してくる様になった。

#### 実施例4

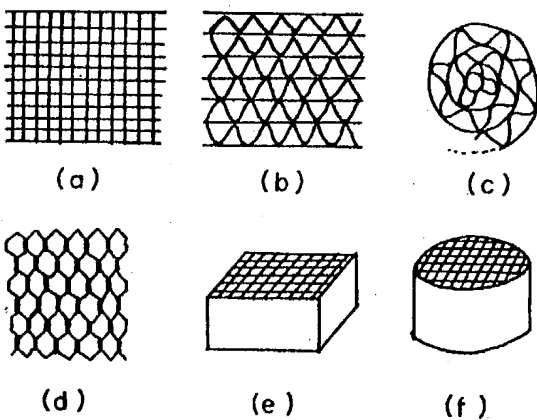
実施例1で用いた活性炭を先ず粉碎し、150~200メッシュのものを選別した後過マンガン酸カリウム水溶液に浸漬し、同様にして活性炭担持マンガン酸化物を得た。一方50~200メッシュの活性アルミナを実施例2と同様の方法でパラジウムを付着せしめた。

細孔断面が一边2.5 $\text{mm}$ の正方形であり、壁の厚みが約0.8 $\text{mm}$ 、全体の形状が75 $\times$ 75 $\times$ 80 $\text{mm}$ の直方体であるプラスチック製ハニカムにエポキシ系接着剤を薄く塗布し、上記活性炭及びアルミナを付着せしめたハニカムを夫々9ヶ宛作製した。実施例3に於ける粒状活性炭担持マンガン酸化物の代りに該ハニカム状フィルタを、又、コージユライト製ハニカムの代りに触媒付着プラスチック性ハニカムを9ヶ宛使用し、実施

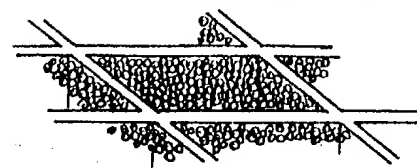
-20-



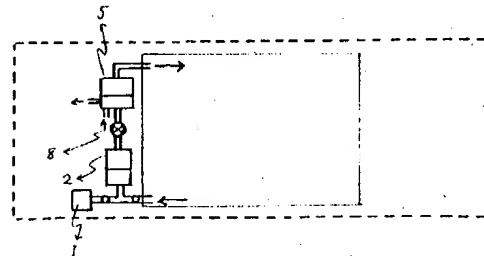
第1図



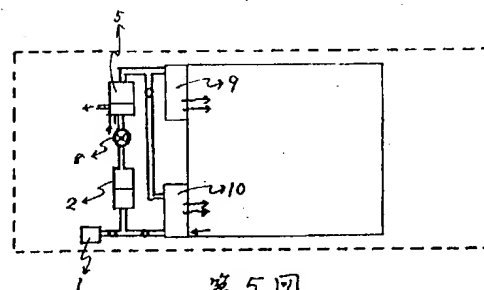
第2図



第3図



第4図



第5図

手続補正書(自発)

昭和48年10月7日

6. 添付書類の目録

- |     |      |    |
|-----|------|----|
| (1) | 明細書  | 1通 |
| (2) | 図面   | 1通 |
| (5) | 願書副本 | 1通 |
| (4) | 委任状  | 1通 |

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自動車室内空気の浄化方法及び装置

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5. 補正により増加する発明の数 なし

6. 補正の内容

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